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Low-Cost Flexible ZnO Microwires Array Ultraviolet Photodetector Embedded in PAVL Substrate

Xinyu Sun¹, Fahad Azad², Shuangpeng Wang³, Lingzhi Zhao^{1,4} and Shichen Su^{1*}

Abstract

With the advent of wearable device technology, fabrication of inorganic semiconductor devices on flexible organic substrates is of great interest. In this paper, a fascinating method and a low-cost flexible substrate material polyvinyl alcohol (PVAL) have been utilized to embed ZnO microwire (MW) array to produce ultraviolet (UV) photodetector (PD) with decent photoresponsivity. The flexible PVAL substrate is relatively cheap and has better bendability as compared to polyethylene terephthalate (PET) and other traditional flexible substrate materials, which makes it unique in comparison to traditional devices. The device shows a current photoresponsivity of 29.6 A/W in the UV spectral range (350 to 380 nm) and maintains an excellent detection performance with even a bending angle of 180°. In dark, a low current of 1.4 μ A at 5 V bias and response time of 4.27 ms was observed. In addition to the excellent device performance at wide bending angles, the fabricated device also performs well with the bending radii close to 0. Therefore, ZnO MW array PD has a great potential for the real-time monitoring of harmful UV exposure to warn the users for the appropriate arrangement avoidance.

Keywords: ZnO microwire array, Ultraviolet photodetector

Background

The detection of ultraviolet (UV) light is important in fields like astronomy, environmental monitoring, and in many biological processes [1]. UV light exposure causes mutation in p53 tumor suppressor genes that causes skin cancer [2]. Therefore, to prevent the injurious effects of sunlight on human health, proper monitoring of these radiations is essential. Furthermore, it is more appropriate to monitor individual's UV exposure as the amount of sun exposure varies from person to person [3]. With the advent of wearable technology, users can now monitor the UV exposure in real time and they can also receive alerts regarding surrounding radiation conditions and their biometric parameters [4]. Therefore, a wearable device with flexible UV PD that could render efficient detection under the bending conditions (required for performing daily

activities of user) is essential to monitor individual's UV exposure.

ZnO, a typical II–IV direct-gap semiconductor, has a wide bandgap (3.37 eV at 300 K) and large excitation binding energy (60 meV). It is one of the most compatible materials for photonic applications such as UV photodetectors and light-emitting diodes (LEDs) [1, 5]. The dominant crystalline structure of ZnO is hexagonal wurtzite with spontaneous polar angle along the *c*-axis, which has been observed in various ZnO nanostructures such as thin film [6, 7], nanorods [8, 9], nanowires [10, 11], nano tetrapods [12, 13], nanobelts [14, 15], and nanocombs [16, 17]. Patterning and alignment of these nanostructures is crucial for device fabrication [18]. To align nanorods and nanowires, several methods such as horizontal manual alignment [19, 20], dielectrophoresis [21, 22], and self-alignment [23] have been explored. Regardless of distinctive properties of these nanostructures, large scale production of these devices is limited due to one by one manufacturing process. The growth of ZnO films using cheap and simple methods has

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attracted the interest of many researchers [24]. Usually, nanostructures of ZnO are fabricated by both chemical and physical vapor deposition methods. Many advanced techniques such as chemical vapor deposition (CVD) [25], metal organic chemical vapor deposition (MOCVD) [26, 27], pulsed laser deposition (PLD) [28, 29], radio-frequency magnetron sputtering (RFMS) [30, 31], and electron beam evaporation (EBE) [32, 33] have been utilized to grow ZnO films. Wet chemical method such as Sol-gel deposition has also been utilized with several casting methods such as dip [34], spin [35], and spray coating to grow ZnO. Sol-gel is an inexpensive and simple method for large scale and roll to roll production. All the discussed method provides ZnO films with large surface area which needs further patterning to meet design requirement of the device. For the patterning of these devices, slow process like photolithography is used [36]. Moreover, etching constituents which are used for patterning are not compatible with the flexible substrate in some cases [37].

Other manufacturing approaches have also been utilized to prepare on-demand ZnO pattern shapes. Some novel approaches have proven to be limited in terms of device cost and performance [26, 32]. Polycrystalline ZnO with a huge amount of grain boundaries fabricated by electrospinning was found to effectively reduce dark current and significantly increase photoresponsivity. Generally, there are two types of PD: photovoltaic PD and none-junction/metal-semiconductor-metal (MSM) PD [19]. Photovoltaic PD has two types: Schottky and P-N/PIN junction [38], whereas MSM PD has a much simpler structure and fabrication process as compared to photovoltaic PD. Therefore, MSM PDs are preferable in practical applications and it is worthwhile to investigate the factors which improve the performance of these detectors [39].

The selection on flexible substrate of ZnO UV PD is crucial to device performance too. According to the variety of nanostructures, shapes and sizes, and synthesis methods, ZnO has been synthesized on diverse substrates in the literatures. Polyethylene terephthalate (PET) and poly urethane (PU) were frequently utilized for flexible ZnO UV devices. Zhang et al. fabricated a ZnO UV PD based on flexible PU fibers; however, the device has worse performance of low photocurrent attributed to the rough surface of woven PU fibers [40]. In some ZnO nanowire UV PD, ZnO nanowires need to be synthesized directly on the substrate in a furnace with extremely high temperature. But almost every organic flexible substrate cannot endure the high temperature of own to low melting point. As a result, reasonable device structure and selection of flexible substrate material lead to the performance of flexible ZnO UV PD.

In this study, a ZnO MW array embedded in soft PVAL substrate has been demonstrated to be an effective UV

PD. We used liquid PVAL glue to cover the most part of ZnO MW array except the surface of hexahedron structure of ZnO MWs. The PVAL glue was then dried, and Au interdigital electrodes were deposited. This PD device has an excellent flexibility and bending strength, which is proven by its ability to work under large bending angles and bending radii for multiple cycles. This PD device was found to have a rapid response time of 4.27 ms and high photoresponsivity of 29.6 A/W. Thus, it is an excellent candidate for wearable devices to monitor UV exposure in order to reduce the possible health hazards.

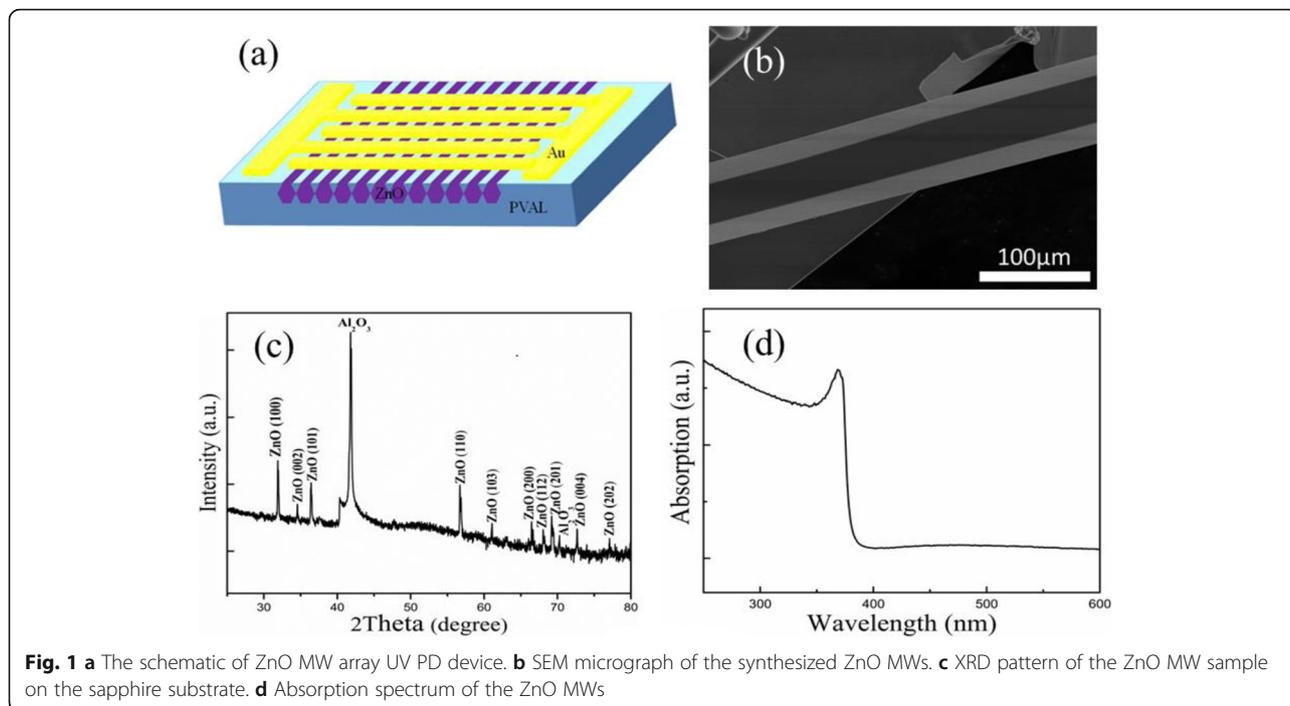
Methods/Experimental

A schematic of the ZnO MW array UV PD is presented in Fig. 1a. The diameter of MWs is 40–50 μm . The MWs were grown by the chemical vapor deposition (CVD) technique. 99.99% powder of Zn sintered to 980 $^{\circ}\text{C}$ for 1 h and turned to gas of Zn in N_2 ambient, introduced O_2 and stayed 980 $^{\circ}\text{C}$ for 1 h, and cooled down to room temperature and got ZnO MWs; more experiment details could be taken in our previous work [41]. ZnO MW arrays of large diameter (40–50 μm) and length (3–5 mm) have been utilized in Fig. 1b for this study. The glass substrate was washed by acetone, ethanol, and deionized water successively. The ZnO MW array was then moved to the glass substrate and compelled to adapt to the surface of glass substrate. PVAL glue was then added dropwise (1 ml) on the ZnO MW arrays evenly. The substrate with ZnO MW array was then kept into drying oven (60 $^{\circ}\text{C}$) for 1 h. The ZnO MW array structure was then peeled off from the glass substrate. Au interdigital electrodes with five pairs of electrode fingers (the gap between adjacent electrodes is 100 μm , finger length is 200 μm) were then deposited on the ZnO MW arrays and PVAL substrate to complete the device fabrication. Figure 2 could explain the fabrication of this photodetector device briefly. This configuration protects ZnO MW arrays as they are embedded in the flexible PVAL substrate. Only the surface of these MWs was exposed to make contact with Au interdigital electrodes.

The morphology and structure of ZnO MWs was characterized by scanning electron microscope (SEM, ZEISS Gemini 500), optical microscope, and X-ray diffractometer (XRD, BRUKER D8 ADVANCE Germany). The absorption spectrum was obtained using a continuous He–Cd (325 nm) laser as an excitation source. The room temperature electrical and photoresponse properties of the fabricated device were measured with a semiconductor characterization system (Agilent B2901A).

Results and Discussion

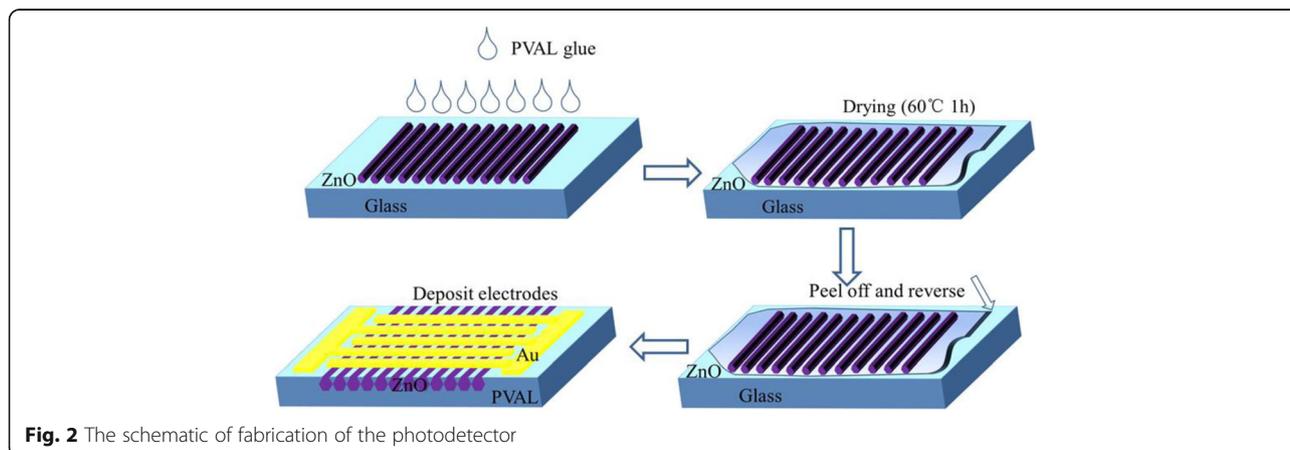
Figure 1b represents a typical SEM image of the synthesized MWs. The MWs were found to have diameters of 40–50 μm and lengths of several millimeters. The XRD



pattern of the ZnO MWs in Fig. 1c indicates wurtzite structure; no secondary phase was detected in the XRD pattern [42]. The absorption spectrum of the as-prepared ZnO MWs is depicted in Fig. 1d indicating good crystallinity with low deficiencies [43].

Figure 3 shows fabricated ZnO MW array PD with no bending (Fig. 3a), 90° bending (Fig. 3b), and 180° bending (Fig. 3c). Figure 4 shows I–V characteristics of ZnO MW array PD devices with and without UV light illumination, 90° bending, and 180° bending. The linear behavior indicates ohmic contact due to lower work function of ZnO (4.5 eV) compared to that of Au (5.1 eV) [44], thus leading to the band distortion and the formation of the depletion region adjacent to the

interface. Once the junction is illuminated by the UV light (380 nm), the electrons and holes generated within the depletion region, immediately move in opposite directions by the built-in potential that gives rise to generation of circuit current. The current increased dramatically, which illustrates that the flexible PD possesses high sensitivity. The flexible UV PDs usually have lower photocurrent compared to the traditional PDs based on Si/SiO₂ substrate due to the poor contact between the material and flexible substrate. But in this study, the embedded ZnO MW arrays have excellent contact with PAVL substrate which is shown from the high sensitivity. The Fermi energy level in ZnO is higher than that of Au. Therefore, electrons will diffuse from



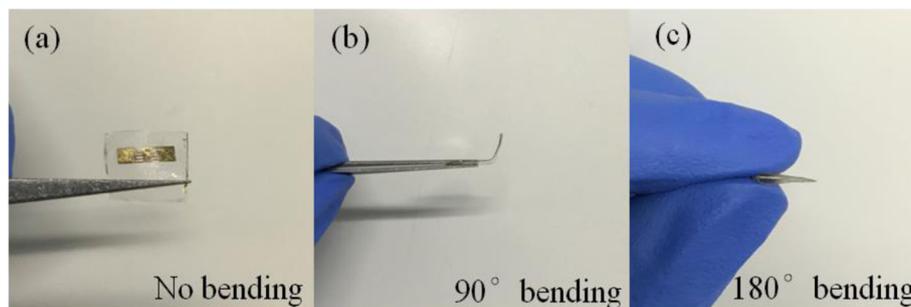


Fig. 3 The schematic of ZnO MW array PD when there is **a** no bending, **b** 90° bending, and **c** 180° bending

ZnO side to Au and a potential barrier will be established that will oppose the further electron flow across the Schottky junction. When an external strain is applied, it creates a negative piezopotential at the interface of Schottky junction which forces electrons to move away from the interface. The repulsion of electrons from the interface will further deplete the interface and increase the height of potential barrier. Although increase in barrier height and width is suitable for the photoexcited extraction and separation, it will alter the transport behavior due to piezoresistance effect. However, the change in the transport behavior is a symmetrical effect which only alters the resistivity of the semiconductor not the interface properties. In this work, the charge transport process due to unsymmetrical variation of current under negative and positive bias is dominated by piezoelectric effect. Hence, the photocurrent decreases with the increase in bending angle.

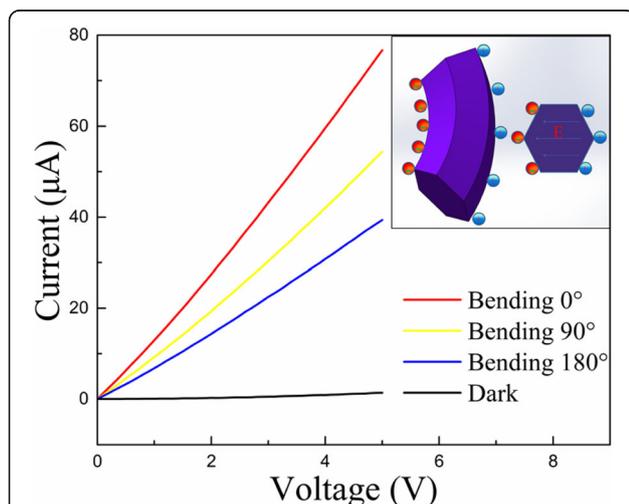


Fig. 4 I–V characteristic in dark and under UV illumination at different bending angles. The inset (left) shows the bending strain-induced nonmobile ionic charges at the outer (positive) and inner (negative) surfaces of the ZnO MW, and the inset (right) shows the piezo-induced electric field and piezopotential distribution at the cross-section of the bent ZnO MW

Wang et al. has discussed the effect of piezoelectric effect on electronic transport properties of ZnO nanowires (NWs) [45]. The positive and negative charging of outer stretched (positively strained) and inner compressed (negatively strained) surface, respectively, in a bent ZnO NW were assigned to be the reason of change in IV characteristics (Fig. 4 inset). The induction of these static ionic charges happens due to the piezoelectric effect. The piezoelectric field along the NW is given by $E = \varepsilon/d$, where ε and d are strain and piezoelectric coefficient, respectively. Two mechanisms were postulated to describe reduction in conductance of NW: (i) the effective carrier density of ZnO NW lowers as the free electrons trap at the inner arc and ions on outer arc surface of the bent NW; (ii) the reduction in the width of conducting channel due to repulsion of electrons across the width, by the induced piezoelectric field.

In our work, the soft layer of PVAL in this MW array UV PD device plays a crucial role in electronic transport. The electron trapping at the interface states builds up depletion region inside the MWs which results in decreasing the effective channel area and creating the surface potential barrier ϕ_s between the MWs and the PVAL dielectrics. When ZnO MW UV PD devices are bent, the electron trapping at the interface states is influenced by different charged surfaces induced by the piezoelectric effect, resulting in the change of transport characteristics.

In the unbent ZnO MW UV PD, the trapping of electrons produces ϕ_s and the band bends upward. When external force is applied to bend the ZnO MW array PD, the applied strain also bends the ZnO MWs. The bending of MWs induces piezoelectric potential ϕ_{PZ} due to movement of Zn^{2+} ions away from O^{2-} ions. The effective potential at the interface varies due to the effect of ϕ_{PZ} on ϕ_s by altering electronic transport properties of ZnO MW array PD due to variation in electron trapping. Negative charge appears on the compressed side of ZnO MW which reduces electron trapping due to repulsion on this side. Whereas, stretched ZnO MW side has positive charge which enhances the trapping of free electrons.

A red shift in the photoresponse wavelength (Fig. 5) was observed by decreasing the bending angle. First principle DFT simulations have been performed on this ZnO MWs under pure tensile and compressive strains to evaluate strain-induced change in the bandgap [46]. For these simulations, the ZnO MWs were strained axially. All the structural optimizations and energy calculations were performed based on pseudopotentials with localized atomic orbital basis sets within the Perdew-Burke-Ernzerhof general gradient approximation implemented in the code SIESTA [47, 48].

To obtain a relationship between the bending angle and bandgap, bandgaps at different bending angles were measured; the data is shown in Fig. 6. The bandgap can also be calculated as a function in the framework of a six-band effective-mass envelope function theory [49]. A significant reduction in the bandgap was observed with decrease in the bending angles. The bandgap decreases from 3.37 eV (bulk) to 3.29 eV due to increase in bending angle from 0° to 180°, respectively, which is in agreement with the six-band effective-mass envelope function theory.

The bandgap and resistance of these ZnO MWs were changed with bending along with photocurrent and the photoresponsivity of the ZnO MW array UV PD also changed. Figure 7 shows the spectral photoresponsivity of ZnO MW array UV PD at different bending angles. It is apparent that the photoresponsivity decreases with the increase in bending angles. The photoresponsivities were measured to be 29.6A/W, 17.1A/W, and 0.95A/W for the bending angle of 0°, 90°, and 180°, respectively. Although, the external stress reduces the photoresponsivity of ZnO MW array UV PD, but even at bending angle of 180°, it is still responsive to UV radiations. Furthermore, the

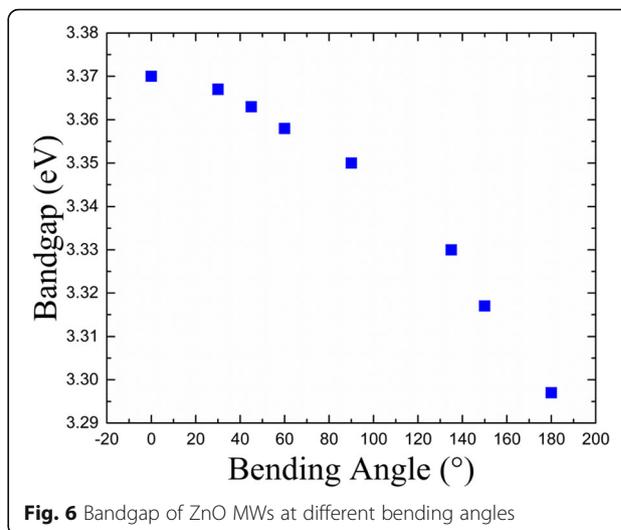


Fig. 6 Bandgap of ZnO MWs at different bending angles

photoresponsivity of ZnO MW array UV PD device were recovered on unbending the device.

Figure 8 presents the dependence of decay times on bending angle for the ZnO MWs array PD device. 266 nm Nd:YAG pulsed laser was used to illuminate the PD device for 30 ns (pulse width) and a bias of 10 V was applied. A reduction in the decay time with increase in the bending angle was noticed. The corresponding values for decay time was found to be 6.18 ms, 6.02 ms, and 4.27 ms for the bending angles of 0°, 90°, and 180°, respectively. The rising time was found to be 4.08 μs which is limited by the pulse width (inset in Fig. 8). The reduction in the decay time can be explained by considering the band diagrams of these MWs for unbent and bent cases. A space-charge depletion layer exists at the surface of these n-type ZnO MWs, and fermi energy

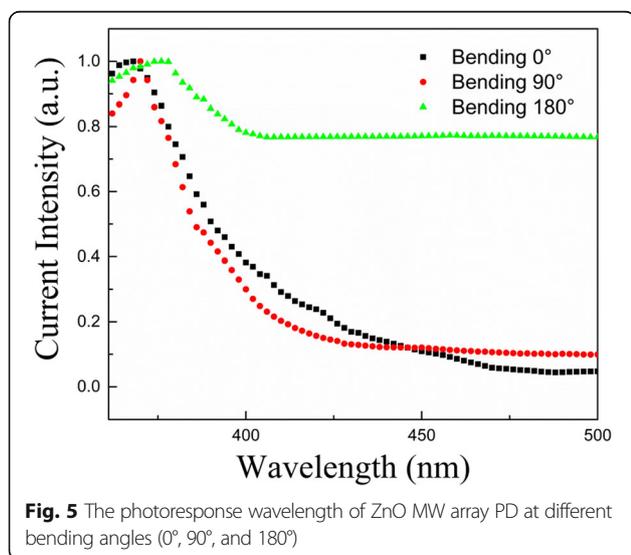


Fig. 5 The photoresponse wavelength of ZnO MW array PD at different bending angles (0°, 90°, and 180°)

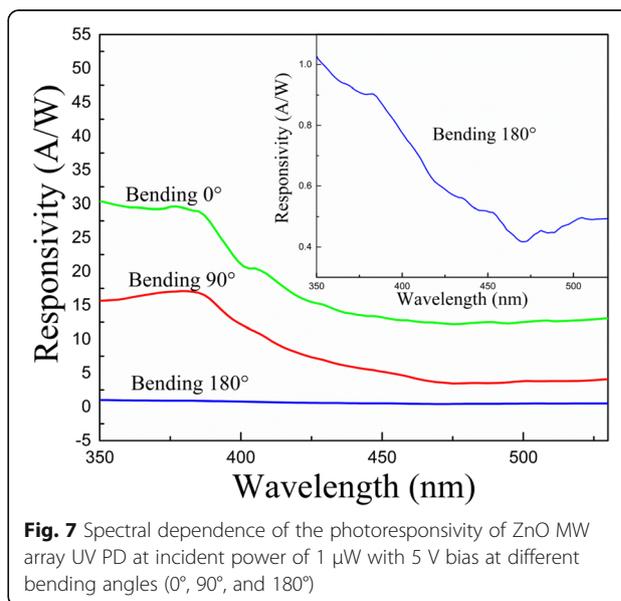
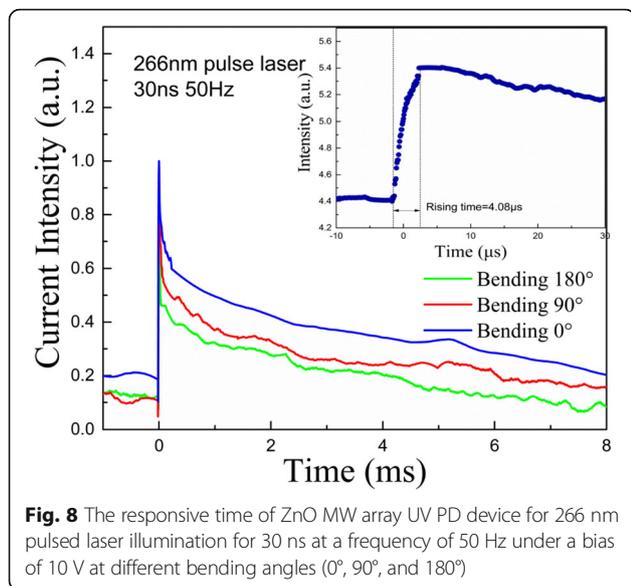
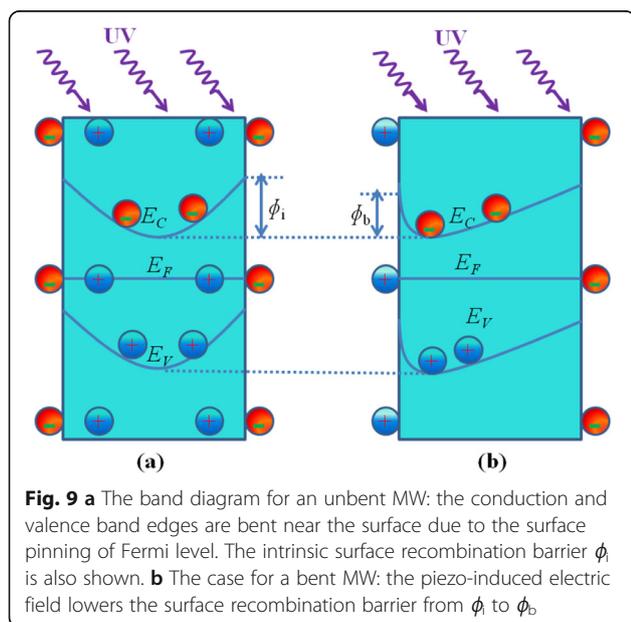


Fig. 7 Spectral dependence of the photoresponsivity of ZnO MW array UV PD at incident power of 1 μW with 5 V bias at different bending angles (0°, 90°, and 180°)



level pins between the forbidden gap at the surface [50, 51]. The depletion layer width depends upon MW's thickness and its atmosphere and doping level which can be controlled by manipulating these factors. In the unbent ZnO MW, conduction band edge (E_c) and valence band edge (E_v) bend upward near the surface of MW and the space-charge depletion region extends up to E_c and E_v band, as shown schematically in Fig. 9. Therefore, photoexcited holes migrate to the surface and electron prefers to stay in the inner part of the MW. The high surface to volume ratio of MWs plays an important role in easy trapping of holes at the surface. The trapping of carriers in surface traps is the dominant



recombination mechanism [52]. The separation between photoexcited electrons and holes reduces the recombination of non-equilibrium carriers. Therefore, to recombine with holes at the surface, electrons have to cross a potential barrier ϕ_i (Fig. 9a). When surface recombination controls decay time of the persistent photocurrent, the recombination rate is given by $\exp(-\phi_i/kT)$ [52].

For the bending case, the induced piezoelectric field modifies energy bands. At the negatively charged surface of the MW, E_v moves towards while E_c moves away from Fermi level. Whereas, near the positively charged surface, both E_v and E_c moves closer to Fermi level, as shown in Fig. 9b. The intrinsic recombination barrier ϕ_i (Fig. 9a) for the unbent case is higher than that of the potential barrier ϕ_b for the bending case (Fig. 9b). Therefore, the recombination rate increases due to reduction in the ϕ_b upon bending. The decay times for the bending case also get shorter as it depend upon the recombination barrier.

Conclusions

In this work, the fabrication of ZnO MW array flexible UV PD embedded in PVAL soft substrate was demonstrated. The process is easy and inexpensive. Good ohmic contacts were created between the Au electrodes and embedded ZnO MW array. The highest response time was found to be 4.27 ms and photoresponsivity to be 29.6 A/W for the fabricated device. Degradation of the device was observed under large bending angles and bending radii, but the UV detection performance was not affected significantly. The effect of bending radii on the performance of the device was also studied. The results suggest that the device is compatible for wearable in situ monitoring UV PDs. This process also shows potential for other devices that need flexibility, such as small-size transistors and solar cells for wearable devices. In addition, the simplicity of the fabrication process might support the idea for custom-made devices or in situ fabrication.

Abbreviations

CVD: Chemical vapor deposition; EBE: Electron beam evaporation; MOCVD: Metal organic chemical vapor deposition; MSM: Metal-semiconductor-metal; MWs: Microwires; PD: Photodetector; PET: Polyethylene terephthalate; PLD: Pulsed laser deposition; PVAL: Polyvinyl alcohol; RFMS: Radio-frequency magnetron sputtering; UV: Ultraviolet

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Availability of Data and Materials

The data supporting the conclusions of this article are included within the article.

Authors' Contributions

XS and SS conceived and designed the study. XS performed the experiments. XS wrote the paper. FA, SW, and LZ reviewed and edited the manuscript. All authors read and approved the final manuscript.

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Competing Interests

The authors declare that they have no competing interests.

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